RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 50, NUMBER 8 15 AUGUST 1994-II

Coherent transient cyclotron emission from photoexcited GaAs

Daniel Some and Arto V. Nurmikko

Department of Physics and Center for Advanced Materials Research, Brown University, Box M, Providence, Rhode Island 02912

(Received 23 May 1994; revised manuscript received 5 July 1994)

Femtosecond photoexcitation near, but above, the band gap in GaAs epitaxial layers and doped heterostructures in magnetic fields is shown to produce transient coherent cyclotron emission, lasting for several picoseconds at low temperatures. This emission reflects the relatively long phase relaxation of coherent electronic excitations associated with *free electron* inter-Landau-level transitions. In a doped, wide parabolic quantum well, the photoexcited carriers drive the magnetoplasmon mode of the equilibrium carriers, producing a dramatic resonance as the cyclotron frequency crosses the magnetoplasmon frequency.

The generation of Thz electromagnetic transients from semiconductor surfaces and heterostructures by interband excitation with femtosecond laser pulses is an active topic of research, in part due to the coincidental development of optoelectronic techniques, which allow subpicosecond resolution of the radiated electric field with high sensitivity,[1,2] providing both amplitude and phase. The physical mechanism of the THz generation at the surfaces has been described by two effects, being due either to the acceleration of the photoexcited charges by surface space-charge fields (current surge), or originating from the second-order nonlinearity $\chi^{(2)} \approx \chi^{(3)} E_s$, i.e., optical rectification augmented by the surface field E_s.[2,3] At sufficiently high levels of intensity, it has also been shown how the bulk component of $\chi^{(2)}$ can become important.[4] In undoped quantum-well structures, THz emission has been observed due to coherent beating between light- and heavy-hole excitons[5] and charge oscillations in asymmetric, coupled double quantum wells;[6] the extension of such experiments to superlattice structures has yielded coherent transients interpreted as excitonic Bloch oscillations.[7] Such studies not only demonstrate new sources of THz electromagnetic transients, but are useful in characterizing the properties of the host systems, especially in terms of coherence properties of electronic excitations in quantum structures.

On the other hand, the study of cyclotron emission has been limited to the observation of incoherent generation from free electrons, e.g., by passing a pulsed current through a doped semiconductor in the magnetic field.[8] Such experiments do not afford subnanosecond temporal resolution; furthermore, in actual practice, the associated detection methods preclude phase-sensitive techniques in terms of identifying electronic coherence. In the work described here, we employ THz optoelectronic techniques to coherently detect transient cyclotron radiation (CR) emitted from photoexcited GaAs and GaAs/$Ga_{1-x}Al_xAs$ samples with subpicosecond resolution; the peak power of this radiation is on the order of tens of microwatts. The incident photon energies reach sufficiently above the band gap so that the (coherent) interband excitations are very unlikely to involve excitons; that is, we are focusing our attention on the free carrier, Landau-level excitations. We present results in this paper for two illustrative cases: (a) an undoped epitaxial layer of GaAs (qualitatively similar results have been obtained in undoped GaAs/$Ga_{1-x}Al_xAs$ heterostructures), where the magnetic-field-dependent spectra exhibit simple electron CR behavior whose temporal decay yields a measure of the phase relaxation of inter-Landau-level coherence; (b) a modulation-doped heterostructure (here a wide parabolic $Al_xGa_{1-x}As$ quantum well), which provides the additional presence of a background equilibrium electron gas. In this latter case we show that while photoinduced THz electron-cyclotron emission is also observed, a potent coupling to the equilibrium electrons is seen, producing a striking magnetoresonance.

Both samples were grown in the [100] orientation; in the modulation-doped $Al_xGa_{1-x}As$ parabolic quantum well (PQW) the aluminum concentration was graded across the well from $x=0$ to 0.30 to produce the parabolic potential profile. It is well established for the ideal PQW that the electrostatic potential by the free electrons subtracts from the compositional conduction-band energy profile in the PQW so that a flat band (i.e., field-free region) is obtained across a central portion of the well.[9] However, finite potential gradients do exist in the well region, particularly near the edges, due to the modulation doping and incomplete well filling, further enhanced by deviations from compositional parabolicity. The total equilibrium electron concentration in the 1000-Å-wide well was $n_s = 2.2\times 10^{11}$ cm^{-2} and the mobility at $T=4$ K was $\mu = 10^5$ cm^2/V sec. We chose the PQW structure to contrast with the bulk epilayer, since the effective penetration depth of the incident photoexcitation could be made comparable for the two; furthermore, the doped PQW presents a very good approximation to an ideal three-dimensional (equilibrium) electron slab. The output from a 100-MHz mode-locked Ti:sapphire laser, composed of 70-fsec pulses and wavelength tunable in the vicinity of $\lambda = 800$ nm, was split into two parts, as depicted in Fig. 1. Approximately 200 mW of average power was used to excite the samples, over an area as large as 1 cm^2, maintained typically at $T=4$ K in a superconducting magnet. The second portion of the laser beam was used to gate a photoconductive, silicon-on-sapphire-based antenna. By sweeping the delay of the gating pulse with respect to the exciting pulse, one time resolves the electric field emitted by the sample. The frequency response of the antenna was calibrated by comparing these measurements with a bolometer-based interferometer, which has no inherent bandwidth limitation[10] but poorer sensitivity. The samples were tilted with respect to

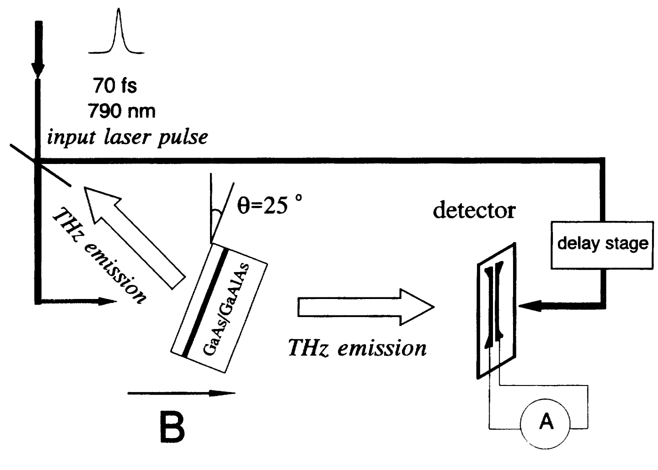

FIG. 1. Experimental geometry of the THz cyclotron emission and detection.

both the B field and the direction of propagation of the excitation beam by an angle of $\theta = 25°$; in this geometry the antenna detected radiation emitted from the sample polarized in the plane of tilt (TM).

In the measurements presented in this paper, the mean photon energy of the exciting laser pulse was chosen to be above the GaAs band gap by about $E \approx 50$ meV (hence also well below the absorption edge of the $Al_xGa_{1-x}As$ barriers outside the PQW section in this sample). Tuning the excitation closer to the band gap did not qualitatively change the results; an abrupt drop in the signal was observed as the photon energy decreased below the band gap, mirroring the absorption edge. The estimated excitation density was equivalent to approximately 10^9 cm^{-2} nonequilibrium electron-hole pairs in terms of real interband excitations. We note that the relatively weak many-electron excitons that have been identified in the modulation doped PQW's are nearly screened at such levels of excitation.[11]

We might anticipate classically, i.e., in a free-carrier transport model, some features of results in the [100]-oriented GaAs wafer, where the velocity **v** gained by a photoelectron or a hole subject to the surface-depletion field will be perpendicular to the sample surface [for the Lorentz force $\mathbf{F} = e(\mathbf{E} + \mathbf{v} \times \mathbf{B})$ to create cyclotron motion, the sample must be tilted with respect to the **B** field]. In the collisionless limit, and ignoring electron-hole (exciton) interactions, the photoexcited carrier will then radiate a circularly polarized wave at the cyclotron frequency $\omega_c = eB/m^*$, where m^* is the appropriate effective mass. Imposing a phase-matching condition for the oscillating carriers across the photoexcited area of the sample produces two spatially coherent beams, which follow the path and divergence of the reflected and transmitted excitation laser beams, as depicted schematically in Fig. 1. Thus we can assume that approximately half of the total emitted radiation is contained in the detected beam, half of which is in the polarization direction picked up by the receiving antenna.

Figure 2(a) shows the electron-cyclotron oscillation wave forms emitted from the undoped GaAs epilayer subject to different magnetic fields, and the corresponding spectra calculated from the Fourier transform of the temporal data (normalized by the known receiver response). The bandwidth of our system ($\nu = 0.2 - 2.0$ THz) naturally focuses on electron-cyclotron motion, rather than hole motion, over the field range in question. We emphasize that Fig. 2(a) plots the electric field, not an intensity, of the emission and is therefore indicative of the coherent component of the radiation from the photoelectrons (in the free-carrier transport model), or interband polarizations over the macroscopic photoexcited area (as discussed below). The electron effective mass deduced from this data is $m^* = 0.068 m_0$, consistent with accepted values for GaAs. The spectral structure in Fig. 2(b) is dominated by the single peaks at ω_c. The large initial increase of the cyclotron emission amplitude with the B field is most likely related to the increase in the inter-Landau-level matrix element, similar to observations of cyclotron resonance in an equilibrium electron gas by steady state far-infrared (FIR) spectroscopy. A dephasing time of $\tau \approx 5$ psec of the electron coherence is obtained by fitting the measured oscillations to an exponentially damped sinusoid $[E(t) = \cos(\omega_c t) e^{-t/\tau}]$. An additional low-frequency component, nearly independent of the B field, is apparent above 1 T; this long-lived, quasistatic component is observable also in the THz emission from various doped and undoped heterostructures. Recent measurements on a different sample indicate that this emission is in fact due to heavy-hole CR, and will be presented in more detail in a later paper.

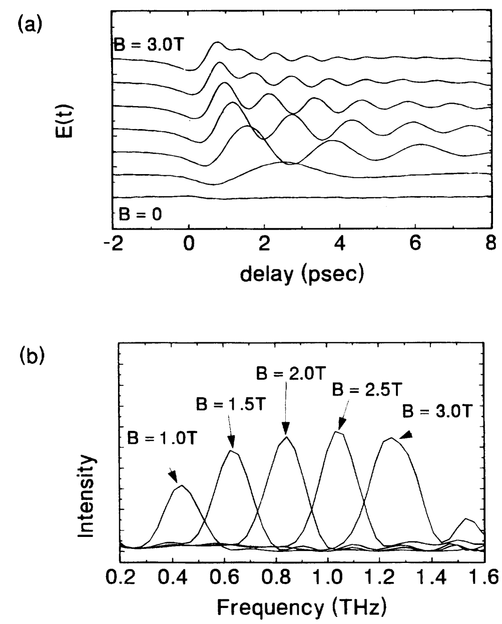

FIG. 2. (a) Transient time-domain wave forms for photoexcited cyclotron emission from a GaAs undoped epilayer at $T = 5$ K in varying magnetic fields from $B = 0$ to 3 T (in increments of 0.5 T), and (b) the corresponding frequency-domain spectra, normalized by the detector response.

Very clear electron-cyclotron resonance emission was also observed in the modulation-doped parabolic-well sample. However, the presence of the equilibrium electrons and the closely spaced subband structure makes the real-time transients more complex, reflecting the coupling of the interband excitations with the equilibrium electrons. The coherence phase decay was approximately $\tau = 3.0$ psec for this sample at $T = 4$ K. The magnetic-field dependence of the Thz emis-

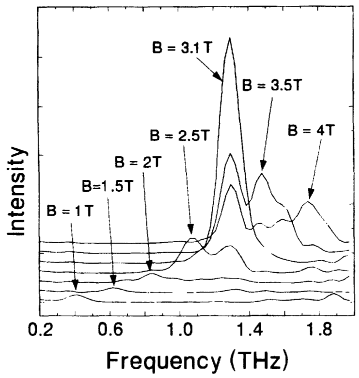

FIG. 3. Spectra of the coherent photoexcited cyclotron emission from the PQW sample in varying magnetic fields, at $T=2$ K. The resonance at 1.3 THz corresponds to excitation of the magnetoplasmon mode of the equilibrium carriers. The spectra have been normalized by the detector response.

sion spectra from the PQW sample is graphed in Fig. 3. Note the distinct resonance enhancement in the emission at $\omega = 1.3$ THz and $B = 3.1$ T. As discussed below, this enhancement runs counter to the observations made by conventional FIR spectroscopy[9] or THz transient transmission measurements,[12] which involve the coupling of the cyclotron and magnetoplasmon modes.

While the observation of the transient cyclotron emission is an intuitively pleasing result from the viewpoint of classically accelerating charges in a magnetic field, a question arises immediately about the long measured dephasing times τ. Supported by a number of time-resolved experiments, it is generally accepted that dephasing times associated with *interband* excitations of GaAs bulk and QW's are below 1 psec in zero magnetic field (including low temperatures),[13,14] and also in high magnetic fields.[15] The THz emission, on the other hand, corresponds to an *intraband* process, and so is governed by dephasing process similar to those at work in cyclotron-absorption measurements: scattering from impurities, phonons, interface roughness, etc.; these may readily match the experimental dephasing times.

In the absence of a rigorous theoretical model for THz generation in a magnetic field, we consider some of the implications of our results here in a speculative spirit. The free-carrier model may be described either semiclassically as ballistic transport (i.e., the unconstrained dynamics of carriers subject to the relevant fields) or quantum mechanically as wave-packet evolution [i.e., the dynamics of a wave packet consisting of excitations of the Landau levels within the laser pulse spectrum (in our case, 10 or more Landau levels are covered by the 20-meV-wide pulse)]. Using the language of ballistic transport, we note that the surface field **E** in the GaAs epilayer is of the order of $10^3 - 10^4$ V/cm, and will tend to accelerate an electron in the direction of the magnetic field (while generating cyclotron motion perpendicular to the **B** field with a drift velocity in the direction of $\mathbf{E}\times\mathbf{B}$). Ballistically, the electron will attain kinetic energy equivalent to that of one longitudinal-optical (LO) phonon in $t = 0.15 - 0.5$ psec. This time domain is also typical of the LO-phonon emission events by hot electrons in bulk GaAs, thus establishing a time scale for inelastic scattering times in our system. The level of photoexcitation is insufficient for screening of the Frohlich interaction, so that many scattering events are anticipated (at least as long as $\hbar\omega_c \ll \hbar\omega_{\rm LO}$, where $\hbar\omega_{\rm LO}$ is the optical-phonon energy).

The long coherence time for the cyclotron motion perpendicular to this acceleration ($\tau = 5$ psec) would then suggest that the LO phonons, if indeed emitted, must carry no net momentum perpendicular to the surface field in each emission event, in order to prevent a break in the cyclotron phase. Since even at $B = 5$ T (highest fields used in our experiment) $\hbar\omega_c \ll \hbar\omega_{\rm LO}$, this condition would require that the electron distribution function (in phase space) be highly anisotropic along the direction of the surface field. Such a situation could arise in conjunction with the coherent emission of LO phonons, as detected recently by Kütt, Albrecht, and Kurz[16] via femtosecond interband excitation screening of GaAs surface space-charge fields.

Having already noted that a rigorous theory for THz generation in magnetic fields is absent, we suggest that our experimental observations are also consistent with the picture in which the magnetic-field-dependent nonlinear susceptibility $\chi^{(2)}(\omega_1 - \omega_2; \omega_1, \omega_2; B)$, enhanced by the presence of a surface field, is responsible for the generation of the transient, coherent interband polarizations. The magnetic field Landau-quantizes the conduction and valence bands; however, the large effective mass of the hole states together with the anticipated higher intraband scattering rates make the valence-band states less discernible in our experimental field range. Thus the frequency difference term $\omega_1 - \omega_2$ corresponds to the inter-Landau-level difference in the conduction band. (We ignore any magnetoexciton effects, since none were found in the excitation spectra of the THz emission near the band edge of our samples.) The $\chi^{(2)}$ process generates coherent electron polarizations, oscillating at $\omega_c = \omega_1 - \omega_2$ in the conduction band, which then decay by intraband scattering processes. The Landau quantization concentrates significant oscillator strength to the near-band-edge interband mixing elements, so that a $\chi^{(2)}$ process is enhanced for the incident pulse spectrum ($\Delta E = 20$ meV), which is spread over several Landau levels. In this spirit, rough correlation is expected of the coherent polarization decay with the conventional cyclotron-resonance linewidth, and to a lesser extent, also the dc mobility. More precise comparison with the FIR spectra is not possible, since the measurements are applied to an equilibrium, free-electron system in a doped semiconductor. Nonetheless, the cyclotron resonance linewidth in our PQW sample of $\Delta E = 4$ cm^{-1}, measured by Karrai *et al.*,[9] compares well with the decay rate obtained in the transient experiments.

Strictly speaking, the THz emission experiment cannot distinguish between radiation due to ballistic transport and a surface-field-enhanced difference-frequency mixing. However, our observation of a sign change of the THz field upon switching the sample tilt angle from positive to negative values does appear to rule out a bulk $\chi^{(2)}$ process.

The pronounced resonance enhancement at $f = 1.3$ THz in the PQW sample coincides with the magnetoplasmon frequency $\Omega_{\rm plasmon}\cos(\theta)$ due to the equilibrium electrons. This collective excitation has been identified and characterized[9] on a similar PQW sample by standard FIR transmission. As is obvious from Fig. 3, this feature appears as a *resonance* rather than the *antiresonance* (anticrossing) observed for coupled eigenmodes, the latter of which results from the hybridization of the CR and magnetoplasmon modes of the equilibrium electrons in a tilted magnetic field.[9] A resonance of the type in the present case is attributable to a *driven oscillator*; i.e., the coherent inter-Landau-level excitation at $\omega_c(B)=eB/m^*$ is driving the magnetoplasmon mode at $\omega_{\rm pl}$. Assuming these modes possess a homogeneous linewidth due to respective damping rates of τ_c and $\tau_{\rm pl}$, the resultant emission line-shape function from this serial process will contain both resonances in the following form: $P(\omega,B)\sim 1/[\{\omega-\omega_c(B)\}^2+1/\tau_c^2][(\omega-\omega_{\rm pl})^2+1/\tau_{\rm pl}^2]$. By including an additional direct contribution of the photoexcited cyclotron emission, this equation qualitatively reproduces the observed spectra over the measured magnetic-field range, including the double peaks, with $\tau_{\rm pl}\approx\tau_c\approx 3$ psec.

A fundamental question concerns the microscopic mechanism for this excitation transfer. Two main possibilities exist: (i) coupling through the radiation field generated by the photoelectron, and (ii) direct electron-electron interaction between the photoelectrons and equilibrium electrons. We speculate that such symmetry-modifying processes that are required are due to the Coulomb scattering by the photoholes. These issues are part of ongoing investigations.

In conclusion, we have observed ultrafast, coherent cyclotron radiation, which is generated by the femtosecond laser pulses incident on undoped and doped semiconductor heterostructures. The decay of the coherent electron polarizations occurs within several picoseconds at low lattice temperatures. The photoexcited carriers couple to the equilibrium electron gas in a doped parabolic quantum well through the magnetoplasmon mode, driving it to resonant emission at ω_c sweeps through $\omega_{\rm pl}$.

We wish to thank M. Shayegan and R. Beresford for the sample, and J. Ding for helpful discussions. This work has been supported by National Science Foundation Grants Nos. DMR-9112329 and DMR-9121747.

[1] M. van Exeter, Ch. Fattinger, and D. Grischkowsky, Appl. Phys. Lett. **55**, 337 (1989).
[2] X.-C. Zhang and D. H. Auston, J. Appl. Phys. **71**, 326 (1992).
[3] X.-C. Zhang, Y. Jin, K. Ying, and L. J. Schowalter, Phys. Rev. Lett. **69**, 2303 (1992).
[4] P. N. Saeta, B. I. Greene, and S. L. Chuang, Appl. Phys. Lett. **63**, 3482 (1993).
[5] P. C. M. Planken, M. C. Nuss, I. Brener, K. W. Goossen, M. S. C. Luo, S. L. Chuang, and L. Pfeiffer, Phys. Rev. Lett. **69**, 3800 (1992).
[6] H. Roskos, M. Nuss, J. Shah, K. Leo, D. Miller, A. Fox, S. Schmitt-Rink, and K. Köhler, Phys. Rev. Lett. **68**, 2216 (1992).
[7] C. Waschke, H. Roskos, R. Schwedler, K. Leo, H. Kurz, and K. Köhler, Phys. Rev. Lett. **70**, 3319 (1993).
[8] E. Gornik, Phys. Rev. Lett. **29**, 595 (1972).
[9] K. Karrai, H. D. Drew, M. W. Lee, and M. Shayegan, Phys. Rev. B **39**, 1426 (1989).
[10] S. E. Ralph and D. Grischkowsky, Appl. Phys. Lett. **60**, 1070 (1992).
[11] M. Shayegan, T. Sajoto, M. Santos, and C. Silvestre, Appl. Phys. Lett. **53**, 791 (1988).
[12] D. Some and A. V. Nurmikko (unpublished).
[13] P. C. Becker, H. L. Fragnito, C. H. Brito Cruz, R. L. Fork, J. E. Cunningham, J. E. Henry, and C. V. Shank, Phys. Rev. Lett. **61**, 1647 (1988); D. S. Kim, J. Shah, J. E. Cunningham, T. C. Damen, W. Shäfer, M. Hartmann, and S. Schmitt-Rink, *ibid.* **68**, 1006 (1992).
[14] P. C. M. Planken, I. Brener, M. C. Nuss, M. S. C. Luo, S. L. Chuang, and L. Pfeiffer, Phys. Rev. B **49**, 4668 (1994).
[15] U. Siegner, M. A. Mycek, and D. Chemla, *Proceedings of the International Quantum Electronic Conference '94* (Optical Society of America, Washington, D.C., 1994), p. 75.
[16] W. A. Kütt, W. Albrecht, and H. Kurz, IEEE J. Quantum Electron. **28**, 2434 (1992).